



CASE EL/2-22773/A/PCT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE PCT NATIONAL STAGE APPLICATION OF
THOMAS SCHÄFER ET AL
INTERNATIONAL APPLICATION NO. PCT/EP
2003/11637

Group Art Unit: 1794
Examiner: MICHAEL E. NELSON

FILED: OCTOBER 21, 2003
FOR: ELECTROLUMINESCENT DEVICE
U.S. APPLICATION NO: 10/531,780
35 USC 371 DATE: APRIL 19, 2005

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

DECLARATION UNDER 37 CFR 1.132

I, Thomas Schafer, a citizen of Germany and presently residing CH-4410 Liestal, Weidweg 15d, Switzerland, hereby declare:

That I was awarded the degree of Diploma Chemist by Technische Hochschule Darmstadt, in 1991; the degree of PhD Chemist by University Basel, Organic Chemistry in 1994;

That I have been employed by Ciba Specialty Chemicals (formerly CIBA-GEIGY AG), Switzerland, as a Research Chemist since 1997, and presently hold the position of Research Chemist ;

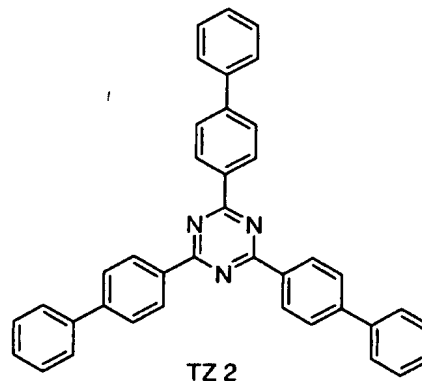
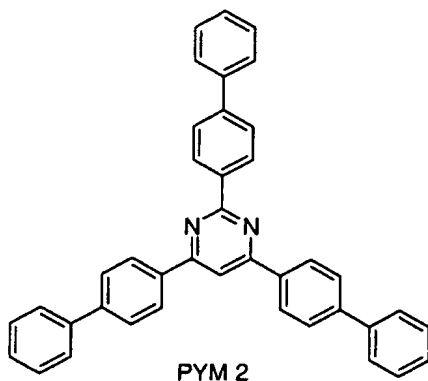
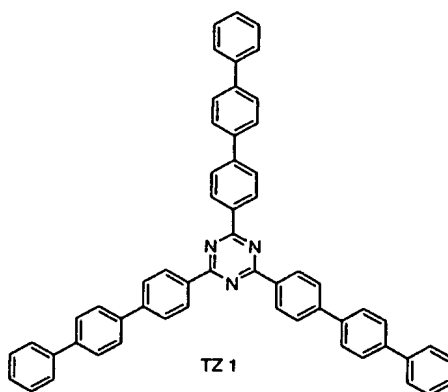
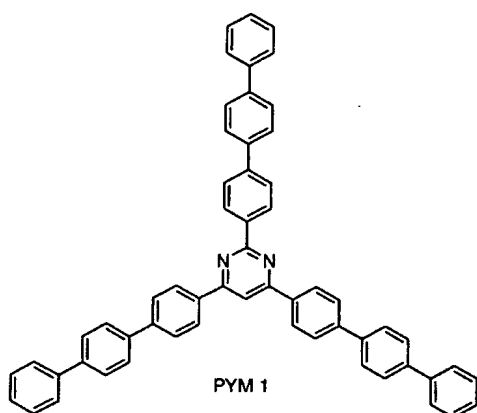
That I have been engaged in the field of organic light emitting diodes for Ciba Specialty Chemicals since 2002;

That I am an inventor on U.S. Patent Application Serial No. 10/531,780.

That the experiments described in the following have been made under my supervision and the evaluation of the results has been done by myself.

Scope of the Study

The experiments herein are designed to directly compare the activity, in the present case the efficiency as an electron transport material, of pyrimidine based compounds of the instant invention to otherwise identical triazine based compounds in an electroluminescent device. Two types of devices are prepared, a red emitting device and a green emitting device. The experiments which follow compare the tris-terphenyl pyrimidine A to the tris-terphenyl triazine B. The compounds were either commercially obtained or prepared following procedures found in the instant specification and were purified by sublimation where necessary.



Device fabrication

The devices used in the experiments comprise the following components:

- a) anode
- b) hole injecting layer
- c) hole transport layer

- d) emissive layer
- e) electron transport layer
- f) cathode.

Devices were fabricated by thermal evaporation in high vacuum ($<10^{-6}$ mbar). The anode a) consisted of ca. 1200 Å of indium tin oxide (ITO) previously deposited on a glass substrate. The cathode b) consisted of 10 Å of LiF followed by 1000 Å of Al. All devices were tested immediately after preparation, without encapsulation, in the nitrogen atmosphere of a glove box (<1 ppm of H_2O and O_2).

In the Red Device, sequentially from the ITO surface:

- b) 600 Å of 2-TNATA (4,4',4''-Tris(N-(naphth-2-yl)-N-phenyl-amino)triphenylamine) as the hole injection layer (HIL),
- c) 300 Å of 4,4'-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (α -NPD) as the hole transport layer,
- d) 300 Å of a aluminum(III) bis(2-methyl-8-hydroxyquinolato)-4-phenylphenolate (BAIQ) doped with 10% of red emitter Ir(piq)2(acac) as the emissive layer.
- e) 30 nm of either PYM 1, PYM 2, TZ 1 or TZ 2 as the electron transport layer.

In the Green Device, sequentially from the ITO surface:

- b) 80 Å of molybdenum oxide as the hole injection layer (HIL),
- c) 300 Å of 4,4'-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (α -NPD) as the hole transport layer,
- d) 300 Å of a 4,4'-bis(carbazol-9-yl)biphenyl (CBP) doped with 6% of green emitter Ir(ppy)₃ as the emissive layer, plus 100 Å aluminum(III) bis(2-methyl-8-hydroxyquinolato)-4-phenylphenolate (BAIQ) as a hole blocking layer (HBL).
- e) 30 nm of PYM 2 or TZ 2 as the electron transport layer.

Experiments and Results

The luminous efficiency, along with the onset voltage (@1000 cd/m²) and maximum luminance measured for the red and green devices are reported in the tables below:

Table 1, Red Device

	ETM (30nm)	C.Eff@1000 cd/m ²	P.Eff@1000 cd/m ²	V@1000 cd/m ²	Max Lum/cd/m ²
1	TZ 1	5.6	2	8.2	6600
2	PYM 1	6.2	2.1	9.1	6700
3	TZ 2	6.4	2.2	9.1	6900
4	PYM 2	7.0	2.2	10.0	5300

Table 2, Green Device

	ETM (30nm)	C.Eff@1000 cd/m ²	P.Eff@1000 cd/m ²	V@1000 cd/m ²	Max Lum/cd/m ²
1	TZ 2	17.6	6.7	8.2	9500
2	PYM 2	20.8	7.2	9.1	5800

Discussion

When used as an electron transport layer, the pyrimidine compounds of the instant invention yield higher luminous efficiency when compared to analogous triazine compounds. This is most striking when measured as C.Eff, but is also seen when measured as P.Eff except for the comparison of PYM 2 with TZ 2 in the red device where the values are the same.

The differences seen demonstrate a significant improvement for the pyrimidine compounds despite the fact that the pyrimidines and triazines both have very similar low lying HOMO values typical for electron transporting materials. For example, HOMO (highest occupied molecular orbital) values calculated from CV measurements in solution using Fc/Fc⁺ as an internal standard revealed a value of -5.6 eV for each of PYM 2 and TZ 2. It appears that the pyrimidine has a much lower lying LUMO (lowest unoccupied molecular orbital) which may be responsible for the improvement.

In any case, given the structural similarities between the pyrimidine and triazine core and the similarities in the low lying HOMO levels, considered a significant factor in electron transport, the differences observed and reported in the above tables are unexpected as well as significant.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements

were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 27th of February 2009


Thomas Schafer